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ULTRASONIC RESEARCH IN THE USSR AND HUNGARY

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Lecture delivered by Paul Gregus at the meeting of the Debrecen section of the Eotvos Lorand Physical Society on 13 April 1951

/The author presents a very good survey of work performed in ultrasonic research, but much of the report seems to have been taken from Der Ultraschall by Ludwig Bergmann, translated into English under the title Ultrasonics and Their Scientific and Technical Applications, published by Bu Ships, Navy Department, as Nav Ships 900,167. However, the author also refers to some of his own research, as well as recent trends in Soviet work./

The catastrophe of the Titanic gave rise to experiments for detecting ~~signaling~~ icebergs moving under the surface of water. At first, so-called sound organs of various types were built which emitted under-water sound signals from the bottoms of the vessels and the echo reverberating from the iceberg was used to determine the distance. The experiments failed, however, mainly because ~~due to the fact that~~ the sounds produced were too weak.

The problem remained dormant until Germany introduced submarine warfare on a large scale during the first world war. The task was the same, namely, how to discover the presence of an object under the surface of water from a great distance. The navies of Britain and France grappled with the problem, but succeeded in solving it only after the Russian Kilovski (1) joined in the effort.

Kilovski's theory was to reverse the piezo-electric effect discovered by the Curies in 1880 to produce high-frequency ultrasonic vibrations ~~emissions~~; and in cooperation with the Frenchman Langevin he laid the foundation of a method which has been the basis of all equipments used for this purpose up to the present. Kilovski died during the war and the equipment was subsequently perfected by Langevin (2). Kilovsky's theory opened a new chapter in ultrasonic research and the object of this article is to discuss the work of Soviet scientists in this field.

Serious theoretical, as well as practical, significance attaches to Sokolov's discovery published in 1929 (3), according to which quartz mosaic does not vibrate [redacted] in the same way as a single quartz plate. Sokolov employed [redacted] Langevin's quartz-mosaic sound generator in his experiments. In essence, the Langevin sound generator consists of quartz plates of identical thickness and shape placed between two steel plates. Since the speed of sound in quartz and steel is practically identical, the whole system vibrates as one piece of quartz with a thickness of $2d + d'$. Sokolov found that the surface vibration amplitude is widely different at different points. When various points with identical amplitudes are connected an arrangement is obtained represented in Figure 1. It follows that the sound space formed by a radiating head is not of uniform ~~does not possess a homogeneous~~ intensity,

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a fact which must be taken into consideration in experiments.

In measuring the relative intensity of sound, a ~~frequency~~ sound-pressure meter ~~published~~ described by Malov and Rzhevkin in 1932 (4) represents another step forward in simplifying this technique. The instrument consists of a thin aluminum lever rotating on a horizontal axis and ~~is equipped on its lower part~~ with a small plate sensitive to sound vibrations attached to its lower end. The deflection of the lever, extending downward into the sound space under examination, may be read on a calibrated scale. In 1933 Malov pointed out (5) that the energy distribution of ultrasonic ~~sounds~~ waves in liquids may be easily determined, even for small intensities, by means of a resistance (thermometer) consisting of a 1.5-~~centimeter~~ cms long and 15 ~~[illegible]~~ μ-thick ~~red-hot~~ iron wire. In 1935, Sokolov recommended (6) that a properly ground ~~Seignet~~ Rochelle salt crystal, which is 150 times more sensitive than the quartz indicators previously used, should be employed as an ultrasonic sound ~~indicator~~ receiver.

Szalay performed pioneering work in 1934 (7) in measuring the speed of ultrasonic ~~sound~~ vibrations in electrolytes and the compressibility of electrolytes. His results were confirmed and further developed by Pasinsky in a paper published in 1938 (8), in which Pasinsky showed that compressibility depends only to a slight extent on the ion radii. Porosorov arrived at a similar conclusion in 1940 (9).

In measuring the ~~absorption~~ attenuation of ultrasonic sound in liquids the intensity of the sound wave is measured at several points of its path directly or indirectly. If the sound wave is not homogeneous, or the source ~~of sound~~ shows even a slight instability in intensity, the measurement is undependable. This uncertainty was avoided by Korolev (10,11) by coupling the measurement with Toepler's simple ~~sliren~~ schlieren method. The picture of ~~the moving~~ a traveling wave shows, in this method, ~~increasing clarity in the direction of the wave proportionate to the~~ a periodic brightening which increases with increasing intensity ~~of the~~ sound at a given point. By ~~means of~~ photometric methods, the observer ~~is in a position to~~ can measure the difference in intensity and thus the sound absorption coefficient between the two points. It is, of course, necessary to use as small sound intensities as possible, because sound intensity is proportional to the ~~deflection of~~ diffracted light only at small sound amplitudes.

Sound absorption measurements in ~~mixed liquids~~ solutions were made by Gurevich and Mikhaylov (12-17), permitting important conclusions as to the structure of liquids. The measurements of emulsions and suspensions made by Vladimirsky and Galani in 1939 (18) solved the problem of sound absorption in a sound space in which the suspended particles do not participate in the movement of the sound space. The ~~empirical~~ experimental absorption values deviated, however, considerably from the values calculated on the basis of classical theory. The ~~empirical~~ experimental values were always greater

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than the values derived from theory. To clarify the problem, Gorodetsky performed measurements and calculations in 1940 (19). He found that the dispersion of ultrasonic waves ~~sounds~~ in liquids is the cause for the discrepancy between the two values. He even discovered a connection between the dispersion of ultrasonic sounds and light.

The property of ultrasonic sounds to modulate light was used by Tumerman and Simanovsky (2) to construct a fluorometer capable of determining ~~by a bell~~ the time required (for the cessation of any kind of) ~~by any~~ luminescence ~~phenomenon~~. Such decay periods ~~The time required to operate the bell is~~ are of the order of 10^{-9} sec. The instrument operates as follows (Figure 3): Quartz Q_1 is caused ~~induced~~ to vibrate at frequency N, which modulates the light originating in source B with ~~in the well-known manner by~~ a frequency 2N. The liquid under examination is in vessel F and is illuminated by the modulated light. The luminescent light rays become parallel after leaving lens L_4 and subsequently pass through quartz Q_2 which likewise has a vibrating frequency N. The sound wave is then microphotographed. Both quartz plates are fed from the same oscillator ~~generator~~. ~~There are~~ two photographs are required to ~~prerequisites for~~ determine ~~determining~~ the decay time ~~required to ring the bell~~. First, the sound wave in the fluorescent light is photographed on a ~~the~~ partly-exposed ~~covered~~ photographic plate ~~by means of fluorescent light~~. Next, the other part of the plate is exposed ~~covered~~ and, after substituting a mirror or white screen ~~shade~~ for the fluorescent substance, another ~~a new~~ picture is taken. The decay time ~~to operate the bell required by the fluorescent light~~ may be calculated from the displacement of the sound wave images ~~pictures~~ on the two photographic plates ~~on the basis of~~ if the modulating frequency is known ~~frequencies~~. The same authors also made use of the light-modulating effect of ultrasonic waves ~~sounds~~ to determine the speed of light in various media.

In investigating the light-modulating effect of ultrasonic waves, ~~sounds~~ Karisimenov pointed out in 1937 (21) that this phenomenon may be utilized in television and communications ~~light telephony~~ instead of the Kerr cells. A great advantage of this process in comparison with the Kerr cells consists in the simplicity of the optical equipment and the high efficiency ~~great strength of light~~. In contrast to the high ~~tension and~~ power required by the Kerr cells (100 watts), ~~a~~ much lower voltages and powers are ~~tension is~~ sufficient (20 volts and 0.05 - 0.2 watts). Another advantage is that the equipment may also be used for ultraviolet light ~~areas~~, whereas the nitrobenzene ~~nitrobenzol~~ used to fill ~~for charging~~ the Kerr cells is not suitable for this purpose. The disadvantage of the process is that it is suitable only for frequencies determined by the quartz which serves as sound source.

As early as 1929 (3), in investigating the speed of ultrasonic sound in solids, Sokolov ~~was the~~ first ~~to~~ pointed out that, since the propagation of ultrasonic waves ~~sound~~ in substances containing cracks is much poorer ~~worse~~ than in homo-

geneous solids, ultrasonic ~~sound~~ waves could be employed for the examination of in-
dustrial materials. The problem deserved particular attention, since it was
theoretically shown that defects of the order of 10^{-6} could be detected by this
method. This method represented a ~~serious advantage in comparison to~~ considerable improvement over X-rays, which
can detect a defect only when it amounts to 1.5 - 2 percent of the thickness of
the material examined.

Sokolov published the first results of his experiments in 1934 (22) and as
early as 1935 he described a practical equipment suitable for industry (23, 24).
The equipment operated on the following principle (Figure ~~A~~ 2): The sound source
consists of a quartz-mosaic plate placed between steel plates "a" and "b".) The
upper plate "a" is screwed into vessel "c" as cover. The surface of "a" is cov-
ered with a thin layer of mercury which is kept from dripping down by skirt "d".)
The sound is transmitted by the mercury to the material "e" under investigation.
On the other side of the material under investigation, facing the sound source,
a vessel with a plane parallel bottom is placed, containing oil or benzene at a
height of a few millimeters. The parallel light rays emanating from source "F" fall on
the surface of the liquid at an angle and are reflected upon a screen. Depending
on the intensity of the sound waves which penetrate the material under investigation,
light spots of different structure appear on the screen. From this so-called
sonogram, conclusions may then be drawn as to the interior homogeneity of the ma-
terial investigated. Larger pieces are pulled through [redacted] in a vessel
filled with oil a few centimeters under the surface while, at the same time,
ultrasonic waves are transmitted through them from below. When the surface is
illuminated as above, similar sonograms are obtained.

The optical procedure outlined in the foregoing serves, however, merely
general informative purposes. Exact location of the defect can be arrived at only
by an examination point by point. Sokolov also worked out a piezo-electric
ultrasonic indicator for this purpose with ~~the employment of the Seignet~~ Rochelle crystal
~~mentioned above~~. This instrument operates as follows:

The piezo-electric crystal plate begins to vibrate mechanically when sound
waves having a frequency corresponding to its own fall upon it. As a result of the
mechanical deformation due to piezo-electric effect, electric charges appear on the
surface. The algebraic sign of these electric charges changes periodically according
to the sound frequency. It is possible to indicate the alternating ~~tensions~~ emf's by various

methods. Because the ~~tension~~ voltage ~~provided~~ generated by the receiving crystal is proportional to the amplitude produced by the sound wave, while the latter in turn is a function of the sound intensity, the change in intensity caused by inhomogeneity in the substance under investigation can be demonstrated. This piezo-electric effect was used by Sokolov also in the ultrasonic microscope published in 1950, which not only indicates inhomogeneity but also locates it exactly and permits it to be photographed.

In essence, this procedure consists in covering the far side of the quartz plate with a layer sensitive to light, which is illuminated by ultraviolet rays. Under the effect of the ultraviolet rays, photoelectrons are ~~detached~~ liberated from the plate, are subsequently accelerated by electric fields, and are ultimately projected through a magnetic and electric lens system on a screen. Since the ultrasonic field creates a piezoelectric charge on the quartz plate in a distribution corresponding to the object under investigation, the resulting charges liberate a larger or smaller number of electrons from the plate. Since, therefore, the photoelectric emission corresponds to the piezoelectric charge distribution produced by the ultrasonic field, the picture of the object investigated appears magnified on the screen. (25)

The ultrasonic waves are transmitted in various substances at speeds characteristic of the respective substances. Sokolov made use of this phenomenon in 1946 (26) to use measurement ~~by~~ ultrasonic speed for the investigation of chemical processes, ~~as well as for the determination of the mechanism of chemical processes,~~ which ~~it~~ were ~~was~~ impossible or very difficult to investigate by other methods. The papers published by Gorbachev (27) and Severny (28) made important contributions to the clarification of the coagulating effect of ultrasonic waves. Our research group, for example, was able, on the basis of these investigations, to regain the cement dust escaping through the dust stacks of the Tatabanya Cement Works by acoustic methods. (29)

Levshin and Rshevkin showed in 1937 (30) and Polotzky in 1938 (31) that the luminescence phenomena produced (in liquids) by ultrasonic waves can probably be explained by the fact that ultrasonic waves create electric charges. The theory of the phenomena was published by Fraenkel in 1940 (32) and was further developed by Natoson (33). They distinguish two kinds of cavitation, namely, equilibrium and breakage cavitation. The first is observed when the gas pressure of the liquid ~~is released~~ decreases slowly, ~~while~~ thus forming spherical cavities ~~are produced~~. The second kind is produced by a quick change in pressure as, for example, under the effect of ultrasonic waves. It causes a break in the liquid and the formation of lens-shaped cavities. Electric charges are released only under the latter circumstances. In the light of these investigations Soloviyeva's paper (34) may be understood, reporting that under the action of 300 kcs effect of ~~an~~ ultrasonic wave, ~~of 300 kcs~~ dilute ferrosulphate solution is transformed into ferrisulphate. Same as (31)

This article led me to the thought of transforming potassium ferro-cyanide into potassium ferri-cyanide by oxydation by means of ultrasonic waves and to employ the resultant Turnbull ~~Trabull~~ blue color reaction to indicate ultrasonic wave intensity. I performed my experiments with 800 and 1,600 kcs. Although the process was unsuitable for the measurement of intensity, nevertheless it yielded interesting results. Thus, ~~[illegible]~~ at on both frequencies I observed the formation of potassium ferro-cyanide proportional ~~in amount~~ to the length of time of ~~the~~ irradiation. At 1,600 kcs it was also possible to show the presence of hydrocyanic ~~nitro-prusside acid hydrogen~~ acid. The latter phenomenon will be understood when consideration is given to the fact that, as a result of cavitation, the oxygen is activated and that in a dilute solution hydrogen peroxide, nitrogen oxide, and nitrogen dioxide are formed. The latter ~~enter into~~ interact ~~interreaction~~ with water and as a result nitrous acid and nitric acid are formed, as was quantitatively shown by Polotzky in 1947 (35). The appearance of hydrocyanic ~~nitroprusside hydrogen~~ acid is obviously the result of the formation of nitric acid.

The fact that organic chemical compounds containing a benzene ~~benzol~~ ring lose their property to absorb ultraviolet light is ascribed by Elpiner (36) to the cavitation effect of ultrasonic waves. I observed similar results in the visible spectrum during the investigation of dyes derived from benzene ~~benzol~~.

The property of ultrasonic waves to age wines and liqueur may likewise be explained by the oxidizing ~~oxydating~~ effect of these waves, as shown by Protopopov's patent published in 1938(37). According to this patent, irradiation during 2 - 30 minutes at frequencies of 100 - 300 kcs leads to an effect as if the alcoholic beverage had been aged for several years. This thought was further developed by the Fuhrer-Tarnoczy-Tari patent, which accomplished the same effect in all kinds of alcoholic products during a few seconds.

The emulsion-forming effect of ultrasonic waves - the practical application of which was pointed out by Popov as early as 1936 (38) - ~~is~~ is likewise a product of the cavitation effect of these waves in the medium ~~space~~. In Popov's view the dispersion, as well as stability, of margarine emulsions produced by ultrasonic waves is far superior than ~~to those produced~~ by other methods.

Dispersion of solids in liquids - first shown by Rzhevkin and Ostrovsky in 1935 (39) - is also based on the cavitation effect of ultrasonic waves. The lead, zinc, bismuth, and sulphur particles dispersed by them were of the order of 10^{-6} centimeter. They formed a true colloid solution showing a strong Tyndall effect. This method was further developed by Shesyulinsky and Tumansky (40, 41) and later by Tmunasky and Maximova (42) for the production of a dilute suspension of indanthrene dyes.

The dispersion effect of ultrasonic waves was used by Seliakov (43, 44) to improve the effectiveness of ammonia and hydrogenating catalysts.

Sokolov (45) was the first to call attention in 1935 to the fact that after being treated with ultrasonic waves, alloys show a lower melting point, ~~and~~ solidify quicker, and, at the same time, become finer grained.

In general, the ultrasonic waves accelerate the process of crystallization, as shown by numerous Soviet authors, among others by Altberg (46), Belinsky (47), and Berlaga (48). Kapustin's results (49) are of particularly great importance in this field. In this article he renders an account of ultrasonic effect on the crystallization of organic substances. The crystals thus obtained were much harder than those obtained without irradiation. He also showed that not only direct irradiation but also the vibration of the walls of the vessel accelerate crystallization and helps produce harder crystals.

This effect, together with the knowledge of rheopexy, led me to the assumption that the solidifaction of cement [redacted] could be accelerated by ultrasonic irradiation. I have performed experiments along this line with frequencies of 6 Mc and 4 Mc and obtained favorable results. All signs point to the fact that lower frequencies may produce even better results which may then be utilized in industry.

The results enumerated in the foregoing represent, of course, only a small part of Soviet ultrasonic research. I did not mention, for example, the ultrasonic effect on electric conductivity and in numerous other physical, physico-chemical, and colloid-chemical investigations. My object was merely to point out the main research fields which are connected with Hungarian ultrasonic research. A special article should be written on Soviet biological, serological [redacted], and other investigations in the field of hygiene. Due to limitation of space only two examples are given in the following.

Istomina and Ostrovsky (50) obtained by 5 minutes of irradiation 30 percent increase in the germinating property and yield of potatoes, while Davidov (51) obtained with 2 - 10 minutes of irradiation a 45 percent increase in the yield of sugar beet seeds. In other plants, ultrasonic irradiation resulted in ~~a~~ colchicine-like effect, which was ~~experienced~~ observed also by Porpaczy in 1948. Similar experiments are being currently conducted by Professor Daniel Feher at Sopron.

Another interesting result has been published by Elpiner (52). Vaccination by whooping-cough virus ~~which has been~~ inactivated by ultrasonic irradiation ~~with ultrasonic~~ ensures immunity to the disease. A similar effect was observed by Garay and Berenessik, namely, cancerogenous matter loses its cancerogenous property after ultrasonic irradiation.

It is impossible to ~~render~~ a complete account of Soviet ultrasonic research within the limits of a short article. However, it may be seen from the foregoing that the Soviets are leading in ultrasonic research. The Sokolov ultrasonic research brigade working in the Physical Institute of the ~~[illegible]~~ Eotvos Lorand University of Sciences is engaged in utilizing these Soviet experiences for the benefit of Hungary's national economy.

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